Supporting Material for the manuscript

Behavior of oil droplets at the membrane surface during crossflow microfiltration of oil-water emulsions

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by

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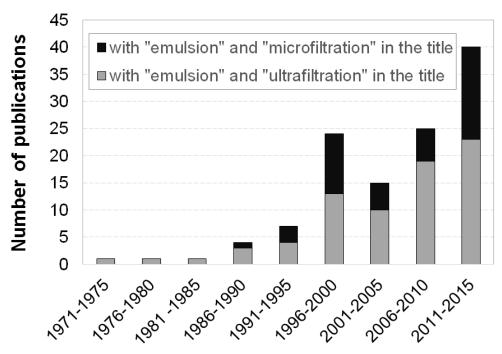


Figure S1: Number of publications with a) both "emulsion" and "ultrafiltration" in the title and b) both "emulsion" and "microfiltration" in the title. Source: Google Scholar. Retrieved: November 1, 2015.

S1. Velocity field in the plane Couette flow

Consider unidirectional (x) flow of an incompressible fluid between two parallel stationary plates as a function of the distance, y, between the plates due to a constant pressure drop. Navier-Stokes equations are simplified to

$$\frac{d^2v_x}{dy^2} - \frac{1}{\mu_t}\frac{dP}{dx} = 0 \tag{S1}$$

Integrating eq. (S1) twice with no slip boundary conditions $[v_x]_{y=0} = 0$ and

$$[v_x]_{y=H} = 0 \text{ gives}$$

$$v_x(y) = \frac{1}{2\mu_1} \frac{dP}{dx} y(H - y)$$
(S2)

The maximum velocity, v_x^{max} , is achieved at the centerline of the flow (y = H/2):

$$v_x^{max} = \frac{1}{8\mu_1} \frac{dP}{dx} H^2 \tag{S3}$$

Average velocity in the channel:

$$\bar{v}_x = \frac{1}{H} \int_0^H v_x(y) dy = \frac{1}{12\mu_1} \frac{dP}{dx} H^2 = \frac{2}{3} v_x^{max}$$
 (S4)

Shear rate is given by

$$\dot{\gamma} = \frac{dv_x}{dy} = \frac{1}{2\mu_l} \frac{dP}{dx} (H - 2y) \tag{S5}$$

and the shear rate at the membrane wall is

$$\dot{\gamma}_{w} = \left[\frac{dv_{x}}{dy}\right]_{y=0} = \frac{1}{2\mu_{l}}\frac{dP}{dx}H = 6\frac{\overline{v}_{x}}{H} \tag{S6}$$

Velocity at $y = r_{drop}$:

$$[v_x]_{y=r_{drop}} = \frac{1}{2\mu_I} \frac{dP}{dx} r_{drop} \left(H - r_{drop} \right) \cong \frac{1}{2\mu_I} \frac{dP}{dx} r_{drop} H = 6\overline{v}_x \frac{r_{drop}}{H}$$
(S7)

and the shear rate at $y = r_{drop}$:

$$\dot{\gamma}_{w} = \left[\frac{dv_{x}}{dy}\right]_{v=r_{drop}} = \frac{1}{2\mu_{l}} \frac{dP}{dx} \left(H - 2r_{drop}\right) = 6 \frac{\overline{v}_{x}}{H} \left(1 - \frac{d_{drop}}{H}\right) \tag{S8}$$

The above derivation is based on the following assumptions:

- 1. The flow is unidirectional and steady
- 2. Gravity can be neglected
- 3. Fluid is Newtonian
- 4. No slip condition at the channel walls

S2. Geometry of an oil droplet pinned at the entry to a cylindrical pore of a membrane.

As can be seen by considering triangles ABC and BCD in Figure S2, the lever arm, ℓ , for drag forces on the droplet around point A and the sine of the droplet's angle of repose, α , are:

$$\ell = \sqrt{r_p^2 + [r^* \cos(\pi - \theta)]^2} = \sqrt{r_{pore}^2 + (r^* \cos \theta)^2}$$
 (S9)

$$\sin \alpha = \frac{1}{\sqrt{1 + \left(\frac{r^*}{r_{pore}}\right)^2 \cos^2 \theta}}$$
 (S10)

where the radius of curvature of the lagging part of the droplet is given by [1]

$$r^* = \frac{r_{pore}}{\cos\theta} \left[\frac{4 \frac{r_{drop}^3}{r_{pore}^3} \cos^3\theta + (2 - 3\sin\theta + \sin^3\theta)}{2 - 3\cos\theta + \cos^3\theta} \right]^{\frac{1}{3}}$$
(S11)

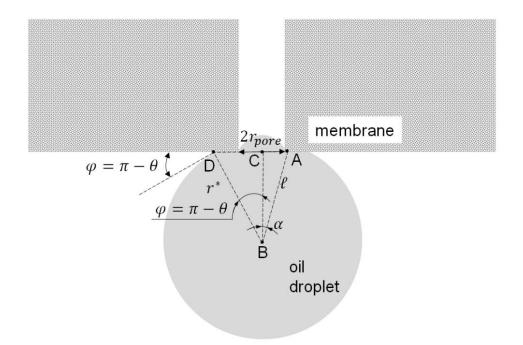


Figure S2: Geometry of the oil droplet pinned at the entry to cylindrical pore of a membrane.

S3. An oil droplet pinned at membrane pore entry: Geometrical considerations

The area covered by one droplet can be calculated based on the values of r^* (eq. (S11); Figure S2) and θ . With this area and the cross-sectional area of one pore (Table 1) known, the average number of pores under one droplet can be calculated (Figure S3):

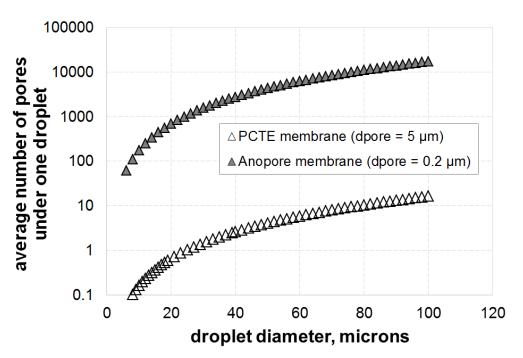
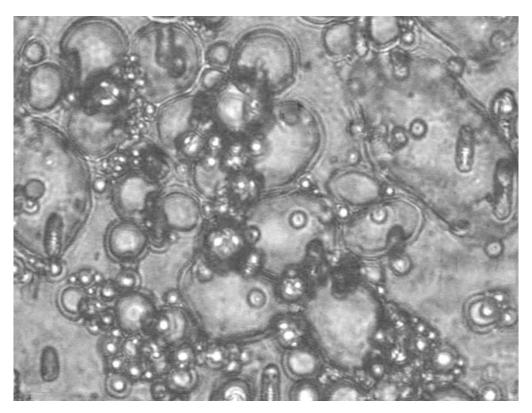


Figure S3: Average number of pores under one droplet.

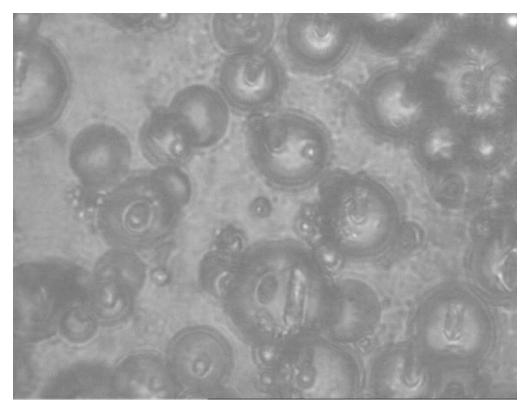
S4. Video evidence of oil droplet behavior at the membrane surface



Video 1: Oil droplet coalescence at the membrane surface.

Video 1 depicts multiple clusters of oil droplets pressing against one another causing the interior oil droplets to deform and become more angular in shape. As the oil droplets attempt to push closer to the pore entrances, it leads to the thinning of the water film between the droplets until the film ruptures and the droplets coalesce. The large cluster of droplets on the top-right of the image will eventually coalesce into a \sim 95 µm droplet that covers multiple 5 µm membrane pores.

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Video 2: Oil droplet permeation.

Video 2 demonstrates the process through which an oil droplet of ~15 μm in diameter can pass through a 5 μm pore in a track-etch membrane. The oil droplet can be seen in the lower-left corner of the image as it attaches to the membrane surface and then slowly squeezes between two other droplets to force its way through the pore. As the oil droplet exits the permeate side of the membrane it can be seen floating towards the top of the image as it leaves the focal plane of the microscope. A sequence of images from this permeation event can be seen in Figure 6 of the manuscript.

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References

[1] F.F. Nazzal, M.R. Wiesner, Microfiltration of oil-in-water emulsions, Water Environ. Res., 68 (1996) 1187-1191.